



## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Application of

Group Art Unit: 1505

Mikio HASHIMOTO, et al.

Examiner: C. Seccuro

Serial No. 08/485,838

Filed: June 7, 1995

For: Propylene Polymer Compositions

Honorable Commissioner of Patents and Trademarks

United States Patent and Trademark Office

Washington, D. C. 20231

Sir:

DECLARATION UNDER 37 CFR 1.132

I, Shin-ya Matsunaga, declare and state that:

1. I am a citizen of Japan, and residing at 1-2, Muronoki-cho, Iwakuni-shi, Yamaguchi, Japan.

In March, 1989, I was graduated from Kyushu University, Faculty of Engineering, Department of Applied Chemistry, and received a Bachelor of Engineering from the same University. In March, 1991, I was graduated from Interdisciplinary Graduate School of Engineering Science, Kyushu University, majoring in Material Science and Technology, and received a Master of Engineering from the same University.

Since April, 1991, I have been an employee of MITSUI Petrochemical Industries Ltd., and till the present time, I

have been engaged in research and development work in Polymer Alloy.

2. I am a researcher of Mitsui Petrochemical Industries Ltd., who is an assignee of the present application.

3. The following Experiments were carried out in order to demonstrate the superiority of the presently claimed process.

#### I

I reproduced Example 1 of Ewen patent. As the result, there was obtained a propylene polymer composition having MFR of about 30 g/10 min., FM (flexural modulus) of 11,200 and HDT (heat distortion temperature) of 95°C. Therefore, if an EPR or PE is further blended therein, FM and HDT would be decreased, and hence, it is expected that resulting polymer blend is inferior in FM and HDT as compared with the data shown in Examples 13, 14, 21 and 22 of the present specification which correspond to the embodiment of the presently claimed invention.

Thus, the process of the present invention provide the unexpected results over the cited reference.

#### II

In order to demonstrate the superiority of the compositions obtained in Examples 13, 14, 21 and 22 reported in the present specification, I conducted the

following Experiments using the metallocene compound catalyst disclosed in Spaleck patent.

[Preparation of Propylene Polymers (A), (B) and (C)]

Propylene polymers (A), (B) and (C) were prepared in accordance with the method for preparing Propylene polymer (7), except for using dimethylsilylene-bis(2-methyl-4-isopropyl-indenyl)zirconium dichloride in place of dimethylsilylene-bis(2-ethyl-4-phenyl-indenyl)zirconium dichloride, and modifying the polymerization time, the amount of hydrogen fed to the polymerization system, etc. so as to obtain the PP's (A), (B) and (C) which have similar MFR's of PP's (9), (7) and (6) respectively.

The properties of the resulting PP's (A), (B) and (C) are shown in the following table 1 together with the data of PP's (7), (8) and (9) used in Examples 13, 14, 21 and 22 of the present specification.

Table 1

	Metallocene	MFR g/10min	( $\eta$ ) dl/g	Mw/Mn	mmn	Tm °C
PP (7)	Me <sub>2</sub> Si(2-Et-4-Ph-Ind) <sub>2</sub> ZrCl <sub>2</sub>	900	0.68	2.02	99.5	-
PP (8)	Me <sub>2</sub> Si(2-Et-4-Ph-Ind) <sub>2</sub> ZrCl <sub>2</sub>	0.75	3.25	2.20	99.6	-
PP (9)	Me <sub>2</sub> Si(2-Et-4-Ph-Ind) <sub>2</sub> ZrCl <sub>2</sub>	13.5	1.64	2.03	99.5	-
PP (A)	Me <sub>2</sub> Si(2-Me-4-IsoPrC-Ind) <sub>2</sub> ZrCl <sub>2</sub>	12	1.7	2.1	-	152
PP (B)	Me <sub>2</sub> Si(2-Me-4-IsoPro-Ind) <sub>2</sub> ZrCl <sub>2</sub>	1165	0.67	2.3	-	152
PP (C)	Me <sub>2</sub> Si(2-Me-4-IsoPro-Ind) <sub>2</sub> ZrCl <sub>2</sub>	1.9	2.65	2.2	-	152

## [Preparation of Propylene Polymer Compositions]

Propylene polymer compositions were prepared in the same manner as in Example 13 of the present specification except for using PP's (A), (B) and (C), and varying the amount thereof as is shown in the following table 2.

The results are set forth in together with the Examples 13, 14, 21 and 22 reported in the present specification.

Table 2

	Ex. 13	Ex. 14	EX. 21	Ex. 22	Comp. Exp. 1	Comp. Exp. 2
PP (7)	50	50	-	-	-	-
PP (8)	50	50	-	-	-	-
PP (9)	-	-	100	100	-	-
PP (A)	-	-	-	-	80	-
PP (B)	-	-	-	-	-	40
PP (C)	-	-	-	-	-	40
EPR-1	15	-	15	-	12	15
EPR-3	-	15	-	15	-	-
PE-1	10	-	10	-	8	10
PE-2	-	10	-	10	-	-
MFR (g/10 min)	9.7	9.5	10.5	10.1	9.3	16
FM (kg/cm <sup>2</sup> )	13,900	13,800	13,500	13,200	11,500	11,900
HDT (°C)	105	104	101	99	88	85

4. From the results of the above experiments compared with the Examples of the present application, and based on my knowledge and experience on Polymer Chemistry, I

The propylene polymer compositions obtained by the processes disclosed in, or suggested from the cited references are inferior in FM (flexural modulus) and HDT (heat distortion temperature) as compared with the polymer composition of Examples 13, 14, 21 and 22 of the present specification which corresponds to the embodiments of the presently claimed invention.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

this 13th day of November, 1996

Shin-ya Matsunaga  
Shin-ya Matsunaga



## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Application of  
Mikio HASHIMOTO, et al.

Group Art Unit: 1713  
Examiner: D.R. Wilson

Serial No. 08/966,652

Filed: November 10, 1997

For: PROPYLENE POLYMER COMPOSITIONS

The Honorable Commissioner of Patents and Trademarks  
United States Patent and Trademark Office  
Washington, D. C. 20231

Sir:

DECLARATION UNDER 37 CFR 1.132

I, Ryoji MORI, declare and state that:

1. I am a citizen of Japan, residing at 1-2, Muronoki-cho, Iwakuni-shi, Yamaguchi, Japan.

In March, 1990, I was graduated from Kyushu Institute of technology, Industrial chemistry of the faculty of Engineering, and received a Bachelor of Engineering from the same university. In March, 1992, I was graduated from Molecular Science and technology of the interdisciplinary Graduated School of Engineering Science, Kyushu University, and received a Master of Engineering from the same University.

Since April, 1992, I have been an employee of MITSUI Petrochemical Industries Ltd., and till the present time, I have been engaged in research and development work in Polymer Science.

2. I am a researcher of MITSUI Petrochemical Industries Ltd. (now, MITSUI CHEMICALS INC.), who is an assignee of the present application.

3. The following Experiments were carried out in order to demonstrate the superiority of the presently claimed invention.

Further to the declaration executed on November 13, 1996 by Mr. Shin-ya Matsunaga, I conducted the following experiments.

#### Preparation Runs

Propylene polymers (D) to (F) were prepared in accordance with the method for preparing Propylene polymer (7), except for varying the amount of hydrogen fed into the reactor, so as to obtain propylene polymers having the following properties.

The properties of PP's (D) to (F) are shown together with the data of PP's (7) - (9) used in Example 13 and 14 of the present specification.

	Metallocene	MFR g/10min	$[\eta]$ dl/g	Mw/Mn	mm	Tm °C
PP (7)	$\text{Me}_2\text{Si}(2\text{-Et-4-Ph-Ind})_2\text{ZrCl}_2$	900	0.68	2.02	99.5	-
PP (8)	$\text{Me}_2\text{Si}(2\text{-Et-4-Ph-Ind})_2\text{ZrCl}_2$	0.75	3.25	2.20	99.6	-
PP (9)	$\text{Me}_2\text{Si}(2\text{-Et-4-Ph-Ind})_2\text{ZrCl}_2$	13.5	1.64	2.03	99.5	-
PP (D)	$\text{Me}_2\text{Si}(2\text{-Et-4-Ph-Ind})_2\text{ZrCl}_2$	-	0.3	2.08	99.6	-
PP (E)	$\text{Me}_2\text{Si}(2\text{-Et-4-Ph-Ind})_2\text{ZrCl}_2$	-	1.5	2.12	99.4	158
PP (F)	$\text{Me}_2\text{Si}(2\text{-Et-4-Ph-Ind})_2\text{ZrCl}_2$	-	0.1	2.00	99.6	-

#### Preparation of Propylene Polymer Compositions

Propylene polymer compositions were prepared in the same manner as in Example 13 of the present specification except for using PP's (D) to (F).

The results are set forth in together with the Examples 13 and 14 reported in the present specification.

	Inventive			Comparative			
	Ex. 13	Ex. 14	Run 1	Run 2	Run 3	Run 4	Run 5
PP(7)	50	50	-	-	-	-	50
PP(8)	50	50	50	50	50	-	-
PP(9)	-	-	-	-	-	50	-
PP(D)	-	-	50	-	-	-	-
PP(E)	-	-	-	50	-	50	50
PP(F)	-	-	-	-	50	-	-
EPR-1	15	-	15	15	15	15	15
EPR-3	-	15	-	-	-	-	-
PE-1	10	-	10	10	10	10	10
PE-2	-	10	-	-	-	-	-
$\eta/\eta$	4.8	4.8	10.8	2.2	32.5	1.1	2.2
MFR (g/10min)	9.7	9.5	81.3	2.6	600	11.9	74.5
FM (kg/cm <sup>2</sup> )	13,900	13,800	14,200	11,800	n.m.	11,500	12,000
HDT (°C)	105	104	107	92	n.m.	88	88

n.m.: not measurable because of bad moldability

4. From the results of the above experiment, and based on my knowledge and experience on Polymer Chemistry, I conclude that:

The ratio of intrinsic viscosity defined in the present claim was essential to attain the objective of the present invention.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be



true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted.

*Ryoji Mori*

this 16th day of February, 1999

Ryoji

Mori